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FOR OFFICIAL USE ONLY**M. G. Politov****Semi-conductor Properties of Colored Crystals of KCl****Chapter I. Color Centers in Alkali Halide Crystals and Phenomena Connected with Them.****I. Introduction**

The study of properties of alkali halide crystals presents a special interest to the physics of solid state, since it allows us to understand some properties of all solid substances.

The coloring of crystals of alkali halides can be produced by different methods, but with this, so-called color centers are formed. Being structure-sensitive property of the crystal, the coloring of the crystals serves as a means for structure study of real crystals.

Colored crystals acquire the properties of semi-conductors. Such crystals luminesce sufficiently well, and addition of admixture of heavy metals makes them suitable for dosimetric purposes.

Alkali halide crystals possess a range of properties which are profoundly influenced by the basic disturbance of the crystal lattice, namely: excitons, electrons and holes, vacancies of the lattice ions, admixture atoms and dislocations. The properties of colored alkali halide crystals are described in a number of works [1-8]. The investigation of coloring phenomena and luminescence considerably widened our knowledge of structure of solid bodies and the mechanism of emission of light.

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A great similarity is found between luminescence and coloring. Therefore, the term "centers", used for denotation of phosphorescence carriers, was used also for denotations of color carriers (absorption) in colored crystals.

The experimental facts have lead to the conclusion that the disruptions in the lattice play a great role in formation of coloring.

Frenkel [7] and Schottky [8] have investigated two types of lattice defects which are now called "Frenkel defects" and "Schottky defects".

The "Frenkel defect" consists of an ion in an interstitial and a vacancy of an ion in the lattice. The "Schottky defect" consists of two vacancies of ions of different signs.

Thermodynamic investigation shows that the free energy of the lattice with vacancies is less than that of a disturbed lattice, consequently the first condition is more favorable. At equilibrium a definite number of ion vacancies corresponds to each temperature. This number grows with increase of temperature. At rapid cooling such a number of vacancies may be "frozen" which corresponds to a higher temperature than that to which the crystal was cooled. Then a metastable condition arises. The "Schottky defects" play the chief role in the phenomenon of coloring alkali halide compounds; the "Frenkel defects" in the silver halides.

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Lately, for explanation of the behavior of real crystals, disturbances of the lattice are produced, called dislocations [9-12].

In the simplest case, the formation of dislocations can be pictured in the following manner. Linearly restricted semi-plane (unfinished plane) is injected between two lattice planes, resulting from which along the borders of the semi-plane forms a disruption of the order of the structural units of the lattice. Thermodynamically it is more probable that the border of the semi-plane (linear dislocation of Taylor-Grovan^{Orowan}) will have ledges, steps, or jogs. F. Seitz [13,14] has pointed out that at plastic deformation these same jogs can evoke the rise of new ionic vacancies which explains the easier coloring of crystals subjected to pressure. Deformed crystals also possess a higher ionic conductivity.

Another type of dislocations are so called spiral dislocations which play an important role at crystal growth. They can be detected in a number of cases on microphotographs on the appearance of spiral-like edges on the surface of crystals.

The present ideas about coloring of crystals are based on the diagram of the energetic levels of the crystal proved by the quantum mechanics [4-6, 15-19].

In the work of Schottky and Stöckmann, it is shown that the division of levels into donor and acceptor levels, based on their energetic position relative to the valence zone and the zone of conductivity is incomplete. From the point of view of

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similar classification, the levels of activation in phosphori, which lie at the filled zone should have the character of acceptors. Meanwhile, the existence of luminescence of the activated crystals is explained to the contrary, by the seizure of electrons from the zone of conductivity onto these levels. The authors show that the complete description of the energetic levels of the centers should consider their situation in the forbidden zone just as their "recharging character" (the change of charge at seizure of the electricity carrier). The authors propose a convenient set of symbols for the description of the condition of admixture centers. There is no actual difference between the character and the functions of the centers in the semi-conductors on the one hand, and in the crystallophosphori on the other. In each separate case, it is necessary to pay as much attention to the position as well as to the recharging character of the levels.

In the zonal theory, the states of the electrons of conduction in the 0th approximation are described by flat modulated waves, while the energetic spectrum of the electrons consists of alternating "allowed" and "forbidden" zones. The zonal theory considers most of phenomena in the 0 approximation, in which the interaction between the electrons and the lattice fluctuations are ignored. At the attempt of explanation of experiment facts, the ordinary zonal theory meets many difficulties. In his monographs, dedicated to the investigations on electron

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theory of crystals, S. I. Pekar denotes two main shortcomings of the zonal theory:

1) Multielectron task is replaced by a monoelectron [one] (the electron is viewed in the periodic potential field), which can not always be substantiated.

2) The interaction of the electrons of conductivity with fluctuations of the lattice is viewed as a minor trouble. In actuality, however, it is great in the majority of crystals [1]. In the mentioned monograph the author expounds on his theory of polarons, i.e. conditions of crystal with a polarizing potential pit, in which an electron is localized; new methods of examination of electrons of conductivity and localized electrons are proposed.

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Deductions or Conclusions, p. 171-174

1. In the colored KCl crystals, besides photoconductivity also electronic dark conductivity is observed.

The dark currents were measured in the field of volumetric charge (in the shortened crystals).

Previous illumination (simultaneously with application of external field) increases the dark currents.

The transition of a colored crystal into "agitated" state at room temperature is connected with formation of R-centers. The dissociation of the R-centers with removal of agitation determines the appearance of the dark electron conductivity.

2. The rise of the dark currents accompanies the formation of complex R- and M-centers in the stained crystals.

The dark electronic conduction of additively colored crystals of KCl depends not only on dissociation of M- and R-centers (the maximums around 80° and 120°C), but also on the dissociation at higher temperatures of F- and K-centers (maximums about 155° and 180°C).

The energy of heat dissociation of R_2 - and R_1 -centers, which was determined by measurements of conductivity with temperature, is respectively 0.31 and 0.43 eV.

3. Production of colloidal K deteriorates the semiconductor

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properties of the colored crystals: dark electronic conductivity decreases and luminescent properties grow worse.

4. The negative volume charge at the surface of the crystal is formed due to rise of excess cation vacancies.

The surplus of cation vacancies can arise: 1) due to the break-up of vacancy pairs at capture of electron; 2) due to departure of cations.

5. In the colored crystals of KCl the electrons do not pass across the border of coloration. At the application of a field to a partially colored crystal a volume charge is formed at the color border. An obvious picture of these phenomena is given by the "cross field" method.
6. For the explanation of the observed "space charge effect" at the electrical puncture of a KCl monocrystal and "photopuncture" of a colored KCl crystal, the results of the study of dark currents were utilized in the field of volume charge.
7. The processes of production and transformation of the centers depends on the structural peculiarities of the lattice.

In the colored crystals the M-centers arise at optical dissociation of the F-centers, if the crystal was irradiated beforehand by severe radiation. This is the proof in favor of the accepted model of the M-center.

8. The decoloration of the KCl crystals in the field evokes

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changes in the lattice conditioning a more intensive darkening at repeated coloration.

9. The K-zone was obtained by several methods in a form manifested by a belt. Up to this time it was observed in the form of a step.

The results of the experiments force to consider as incorrect the explanation proposed by Mott of the rise of the K-zone as the result of transition of electrons of F-centers onto the second level of excitation.

10. In the KCl crystals activated by silver, bands of absorption are observed with maximums at $\lambda = 220, 230 \text{ \AA}$ & $245 \text{ m}\mu$. The coloring evokes the rise of F-band and several bands of additional absorption, out of which the most intensive are the bands with maximums at $\lambda = 285 \text{ \AA}$ & $440 \text{ m}\mu$.

In the presence of F-band a seeming displacement of the maximum from $440 \text{ m}\mu$ to $465 \text{ m}\mu$ is observed. This is explained by the overlapping of the F-band and a new band.

11. The band of absorption with the maximum at $\lambda = 440 \text{ m}\mu$ is identified with the K-band in inactivated KCl crystals.

The rise of the manifested K-band at electrolytical coloring of pure as well as silver activated KCl crystals is accompanied with the rise of colloidal bands of absorption.

12. A proposal is suggested and substantiated that the K-centers

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represent the smallest particles of the alkali metal. The mechanism of formation of the smallest particles of K in the KCl-Ag crystals is proposed at additive coloring by heating in the vapor of K. The role of admixture of silver in the processes of discharge of the smallest particles of K in the net is substantiated.

13. Depending on the regime of coloring of the crystals of KCl-Ag by heating in the vapors of K, a yellow color (K-band) or violet color (F-band) is obtained.
14. Simultaneous gamma-irradiation of crystals of KCl-Ag with different concentration of Ag forms same concentration of F-centers as if the crystals were preheated in vapors of K in such a way that they acquired at this the yellow stain.

In the thermally untreated crystals of KCl-Ag the concentration of F-centers arising at gamma-irradiation is so much less the higher the concentration of silver, which is unprofitable from the point of view of storing of energy in the crystal at irradiation.

In the thermally treated crystals of KCl-Ag not only do the similar F-centers arise independently of the concentration of Ag, but also the concentration of the F-centers arising at gamma-irradiation is higher than in the crystals not subjected to heating.

Pre-coloring of KCl-Ag crystals in the vapors of potassium

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to yellow color facilitates the succeeding coloring by gamma-irradiation. Simultaneous gamma-irradiation of KCl-Ag crystals with same concentration of Ag evokes the rise of numerous F-centers in that crystal which was previously thermally treated.

15. K-centers play a vital part in the phosphorescence phenomenon. KCl-Ag crystals with K-centers show better luminescence properties from the point of their utilization for dosimetry of hard radiation.

The maximal intensity of the stimulated ultraviolet phosphorescence stained by X-rays of KCl-Ag crystals is observed at the wave lengths of light $\lambda = 560 \text{ \AA} \text{ \& } 450 \text{ m}\mu$. These two maximums in the spectrum of excitation correspond to F- and K-bands of absorption.

16. The introduction of admixture of bivalent ions of cadmium into the crystal facilitates the formation of M-centers at additive staining in the vapors.

At electrolytic staining a new band arises with a maximum around $\lambda = 275 \text{ m}\mu$. In the KCl-Cd crystals discolored in the field, this band is more intensive than in the colored crystals.

This band is detected also in pure KCl crystals subjected to discoloration in the field.

A proposition is advanced that the V-type centers are

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responsible for this belt into the structure of which cation vacancies enter.

17. Among the pure KCl crystals subjected to additive coloring and crystals of KCl with admixture Cd the most normal phosphorescence is observed in the case of crystals discolored in the field. It is possible that luminescence of the discolored crystal arises at recombination of electrons with centers of V-type.
18. Additively colored crystals of KCl-Cd phosphoresce weaker than those not subjected to staining.

The explanation of these occurrences has to be connected with the dissociation of complexes "ion of cadmium - a cation vacancy" due to heating at coloring. With the forming of isolated 2-valent ions of cadmium ("positive" charge in the lattice), the electrons recombine without radiation, in accordance with theoretical calculations.

At excitation of uncolored crystals of KCl-Cd, there may take place conversion of 2-valent ions entering into the composition of the complexes into univalent which already are not "positive" disturbance in the lattice.

19. The additive staining worsens the luminescent properties of crystals of KCl with admixture of 2-valent ions of cadmium, but betters the luminescent properties of crystals of KCl with admixture of Ag.

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After the influence of hard radiation the normal as well as the stimulated phosphorescence of KCl-Ag crystals is stronger in that case when an additively stained crystal is subjected to the influence of hard radiation.

20. The storing of energy by KCl-Ag crystals at staining by hard radiation is the reason for the observed stimulated phosphorescence.

The greatest illumination at stimulated phosphorescence is reached at time of photostimulation of 15 - 20 sec.

21. At small doses of gamma-radiation of KCl-Ag crystals the absorption analysis does not detect centers of coloring. However, such crystals show a strong stimulated phosphorescence.

Herefrom it follows that the method of stimulated phosphorescence is a more sensitive method than the method of absorption analysis for the determination of crystal coloring and for the detection of hard radiation.

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